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CATALYST FOR THE CARBONYLATION OF OXIRANES

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Catalyst for the carbonylation of oxiranes

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The present invention relates to the preparation of lactones by catalytic carbonylation of oxiranes in the presence of a catalyst system, to a corresponding catalyst system and to its use.

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The catalytic carbonylation of simple and substituted oxiranes is known per se. The products are often not the desired lactones, or the reaction conditions or the starting materials do not permit efficient preparation or isolation of lactones. The compounds can frequently be obtained by means of complicated and costly syntheses.

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JP-A-09 169 753 describes the carbonylation of epoxides to lactones over $\text{Co}_2(\text{CO})_8$ as catalyst in a flow-through reactor. The conversions are only 30%. This means that a separation and recirculation facility is required to achieve a high yield and purity of the lactone.

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GB-A-1,020,575 relates to a process for preparing polymers of β -lactones. Carbon monoxide and a 1,2-epoxide are reacted to form a β -lactone as intermediate. This is carried out using octacarbonyldicobalt as catalyst. In addition, it is possible to use a promoter selected from among metal halides such as potassium iodide and quaternary ammonium halides such as tetraethylammonium bromide. However, the yields of lactone are less than 10% and the main fractions of the products are polyhydroxypropionic esters. In addition, the reaction is carried out in a complicated manner using a plurality of pressure stages.

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EP-B-0 577 206 relates to the carbonylation of epoxides over a catalyst system comprising a cobalt source and a hydroxy-substituted pyridine compound, in particular 3-hydroxypyridine or 4-hydroxypyridine. The carbonylation is preferably carried out in the presence of a hydroxy compound such as water or alcohols. The activities of the catalysts used are relatively low, and isolation of the lactones is not described. It has also been observed that a change in the reaction mixture occurs after the carbonylation has ended.

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Over a period of 24 hours, polymerization of the lactone takes place. This indicates that the lactone is not unreactive in the reaction mixture. It is also known that lactones can, polymerize in the presence of pyridines.

- 5 Chemistry Letters 1980, pages 1549 to 1552, relates to the reaction of epoxides with carbon monoxide over a rhodium complex as catalyst. The yields are not more than 70%.

J. Org. Chem. 2001, 66, pages 5424 to 5426, describes the synthesis of β -lactones by carbonylation of epoxides over cobalt and Lewis acid catalysts. A system comprising
10 $\text{PPNCo}(\text{CO})_4$ and $\text{BF}_3 \cdot \text{Et}_2\text{O}$ is used as catalyst. The yields are in the range from 7 to 86%. However, the reaction time is from 7 to 24 hours, and the use of large amounts of catalyst is necessary.

J. Am. Chem. Soc. 124, No. 7, 2002, pages 1174 to 1175, describes the preparation of β -
15 lactones by carbonylation of epoxides. The catalyst used is a mixture of a salt of aluminum salts and a tetracarbonylcobaltate. The handling and synthesis of the aluminum compound are complicated, so that the process cannot be carried out on a large industrial scale.

It is an object of the present invention to provide an uncomplicated and efficient process
20 for preparing lactones by carbonylation of epoxides. A further object is to provide a suitable catalyst system for this reaction.

We have found that this object is achieved by a process for preparing lactones by catalytic carbonylation of oxiranes, wherein a catalyst system comprising

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- a) at least one cobalt compound as component A and
- b) at least one metal compound of the formula (I) as component B,



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(I)

where

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- M is an alkaline earth metal or a metal of group 3, 4 or preferably 12 or 13 of the Periodic Table of the Elements,
- R is hydrogen or a hydrocarbon radical which may be substituted on the carbon atoms other than the carbon atom bound to M,

X is an anion,
n is a number corresponding to the valence of M and
x is in the range from 0 to n,

5 with n and x being selected so that the compound is uncharged,
is used as catalyst.

The object of the invention is also achieved by a catalyst as defined above except for the
10 combination $\text{Al}(\text{C}_2\text{H}_5)_3/\text{Co}(\text{acac})_3$.

The catalyst system $\text{Al}(\text{C}_2\text{H}_5)_3/\text{Co}(\text{acac})_3$ has already been described in Die
Makromolekulare Chemie 89, 1965, pages 263 to 268. This reference is concerned with the
15 copolymerization of carbon monoxide with alkylene oxides and does not describe the
formation of lactones.

Lactones are valuable compounds for preparing biodegradable polyesters, cf., for example,
EP-A-0 688 806. These polyesters are widely used, for example as polyols in polyurethane
20 production or as material of construction.

According to the present invention, it has been found that a combination of cobalt
compounds, in particular compounds in a low oxidation state, and metal compounds forms
an efficient catalyst system for the carbonylation of oxiranes to lactones under mild
conditions.

25 In the catalyst system used according to the present invention, preference is given to 0.1 to
1000 mol, particularly preferably from 1 to 100 mol, of component B being present per
mole of component A.

30 The component A is preferably selected so that a cobalt carbonyl compound is present
under the reaction conditions. This means that a cobalt carbonyl compound can be used
directly as component A or it is possible to use a compound which is converted into a
cobalt carbonyl compound under the reaction conditions.

35 R is preferably hydrogen or C_{1-32} -alkyl, C_{2-20} -alkenyl, C_{3-20} -cycloalkyl, C_{6-18} -aryl, C_{7-20} -
aralkyl or C_{7-20} -alkaryl, with substituents being able to be present on the carbon atoms
other than the carbon atom bound to M. R is preferably hydrogen or a monoanionic

hydrocarbonyl group, for example C_{1-32} -alkyl such as methyl, ethyl, i- or n-propyl, i-, n- or t-butyl, n-pentyl or n-hexyl, C_{2-20} -alkenyl such as propenyl or butenyl, C_{3-20} -cycloalkyl such as cyclopropyl, cyclobutyl, cyclopentyl, cyclopentadienyl or cyclohexyl, C_{6-18} -aryl such as phenyl or naphthyl, or C_{7-20} -arylalkyl, e.g. benzyl (the hydrocarbonyl group is preferably alkyl and particularly preferred hydrocarbonyl groups are methyl or ethyl),

X is an anion such as halide (apart from fluoride), sulfonate, oxide, C_{1-32} -alkoxide, amide; with preferred anions being halide or alkoxide, particularly preferably chloride or C_{1-12} -alkoxide,

and n corresponds to the oxidation state OS or the valence of the metal and x is smaller than or equal to n and is not negative (for each oxide ligand, $x = x + 1$).

The component B is preferably $AlCl_xR_{3-x}$ where x is from 0 to 3 and R is C_{1-6} -alkyl. The numbers n and x can be integers or fractions. Fractions can arise in the case of a mixture of such compounds.

If desired, component A or B may also have an uncharged donor L bound in its coordination sphere. A donor L is in general an uncharged compound containing oxygen, nitrogen or phosphorus atoms, e.g. ethers, carbonates, ketones, sulfoxides, amines, amides, phosphines, nitro functions or nitrile functions, etc. Olefins and aromatics are also possible as donors L.

Of course, it is also possible to use mixtures of a plurality of different components B and/or A as catalyst system.

Particular preference is given to a combination of octacarbonyldicobalt and trimethylaluminum or octacarbonyldicobalt and triethylaluminum or octacarbonyldicobalt and tri(sec-butyl)aluminum, or octacarbonyldicobalt and triisopropoxyaluminum.

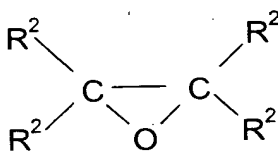
The carbonylation is generally carried out under superatmospheric pressure and at elevated temperature. However, product formation is also observed at a carbon monoxide pressure of one atmosphere. The pressure is generally generated by means of CO gas. In particular cases, the pressure can also be partially generated by means of an inert medium such as argon or nitrogen. The pressures are in the range from 1 to 250 bar, preferably from 10 to 100 bar, particularly preferably from 20 to 60 bar. The reaction can generally be carried out

at from -10 to 200°C. The preferred temperature range is from 20 to 150°C, particularly preferably from 40 to 110°C.

The carbonylation of epoxides can be carried out either batchwise or in a continuous process. It can be carried out either in the gas phase or in an inert reaction medium. This medium is generally a liquid. Suitable liquids are customary solvents such as ethers, diglyme, triglyme, tetraglyme, tetrahydrofuran, dimethoxyethane, hydrocarbons such as hexane, octane, isopar, benzene, toluene, xylene decalin; chlorinated hydrocarbons such as dichloromethane; dichloroethane, dichlorobenzene or polar solvents such as DMF, DMSO, esters, nitriles, nitro compounds, ketones or ionic liquids. Preferred solvents are DME, diglyme, dichloromethane. The oxirane can also be used as reaction medium.

To activate the catalyst system further, it is possible to add donor ligands such as phosphines or nitriles. Application of the catalyst components (e.g. cobalt and alkyl compound) to a particulate support material, e.g. silica or aluminum oxide, makes it possible to carry out the reaction in the absence of solvent as a gas-phase carbonylation.

Suitable oxirane compounds are ethylene oxide and substituted epoxides. These are usually compounds having the formula (II):



(II)

In this formula, the radicals R^2 are each, independently of one another, hydrogen, halogen, a nitro group $-\text{NO}_2$, a cyano group $-\text{CN}$, and ester group $-\text{COOR}^3$ or a hydrocarbon group having from 1 to 32 carbon atoms which may be substituted. The radicals R^2 in a compound (II) can all be the same, some of them can be the same or they can be four different radicals. R^3 can be C_{1-12} -alkyl or aryl.

Use is preferably made of geminally substituted epoxides, particularly preferably epoxides substituted only in the 1 position.

Examples of suitable hydrocarbon groups are C_{1-32} -alkyl such as methyl, ethyl, i- or n-propyl, i-, n- or t-butyl, n-pentyl or n-hexyl, C_{2-20} -alkenyl such as propenyl or butenyl, C_{3-20} -cycloalkyl such as cyclopropyl, cyclobutyl, cyclopentyl or cyclohexyl, C_{6-18} -aryl such

as phenyl or naphthyl, and C₇₋₂₀-arylalkyl, e.g. benzyl. It is also possible for two radicals R² located on different carbon atoms of the epoxy group to be joined to one another so as to form a C₃₋₂₀-cycloalkylene group.

5 Substituents which may be present on the C₁₋₃₂-hydrocarbon group or R above can be, in particular, the following groups: halogen, cyano, nitro, thioalkyl, tert-amino, alkoxy, aryloxy, arylalkyloxy, carbonyldioxyalkyl, carbonyldioxyaryl, carbonyldioxyarylalkyl, alkoxycarbonyl, aryloxycarbonyl, arylalkyloxycarbonyl, alkylcarbonyl, arylcarbonyl, arylalkylcarbonyl, alkylsulfinyl, arylsulfinyl, arylalkylsulfinyl, alkylsulfonyl, arylsulfonyl
10 and arylalkylsulfonyl.

The oxirane compound used is preferably ethylene oxide, propylene oxide, butylene oxide (1-butene oxide, BuO), cyclopentene oxide, cyclohexene oxide (CHO), cycloheptene oxide, 2,3-epoxypropyl phenyl ether, epichlorohydrin, epibromohydrin, i-butene oxide
15 (IBO), styrene oxide or an acryl oxide. Particular preference is given to using ethylene oxide (EO), propylene oxide (PO), butylene oxide or i-butene oxide, very particularly preferably ethylene oxide or propylene oxide or a mixture thereof.

The oxirane compounds to be used for the process of the present invention can be obtained,
20 for example, by epoxidation of terminal olefins using methods known to those skilled in the art. If the epoxidation occurs stereounspecifically, resolution of the racemate has to be carried out. Methods of resolving racemates, e.g. by means of HPLC chromatography using a chiral column material, are known to those skilled in the art. The oxirane compound is advantageously prepared directly in enantiomerically pure or optically
25 enriched form from a terminal olefin by means of established stereoselective processes. A suitable process is, for example, the Sharpless epoxidation (cf. J. Am. Chem. Soc. 1987 (109), p. 5765 ff. and 8120 ff.; and "Asymmetric Synthesis", Edited by J.D. Morrison, Academic Press, New York, 1985, Volume 5, Chapters 7 and 8).

30 Furthermore, optically enriched oxirane compounds can be obtained from terminal olefins or racemic terminal epoxides by the methods described in Jacobsen et al., Tetrahedron Lett. 1997, 38, pages 773 to 776; and J. Org. Chem. 1998, 63, pages 6776 to 6777, which are also simple to carry out on a large industrial scale (see also Acc. Chem. Res. 2000, 33, pages 421 to 431).

35 It is also possible to prepare optically enriched oxirane compounds by mixing an appropriate amount of the racemate with the enantiomerically pure oxirane compound.

As compounds having a terminal double bond, it is in principle possible to use all olefins of this class of compound, e.g. propene, 1-butene, 1-pentene, 1-hexene, 1-heptene or 1-octene.

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In general, the cobalt complexes (A) and the, for example, alkyl compounds (B) are firstly introduced into the reaction vessel either individually, simultaneously or in premixed form, if appropriate with cooling. It is also possible, if desired, for the oxirane compound to be mixed into the solution/suspension of the catalyst components before the latter is transferred to the reaction vessel. Furthermore, the oxirane compound can also be introduced directly into the reaction vessel. The carbonylation is preferably carried out under inert conditions, i.e. in the absence of moisture and air.

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Termination of the reaction and separation and purification of the lactones can be carried out by generally known methods. For example, the lactone can be isolated in a simple fashion by distillation or crystallization.

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The process of the present invention enables 3-hydroxypropiolactones to be obtained from the corresponding enantiomerically pure oxirane compounds. If these oxirane compounds are used in optically enriched form, lactones in which the degree of optical purity corresponds directly to the degree in the oxirane are obtained. Lactones prepared in this way can be used to establish the thermoplastic property profile of the biodegradable polymers, whose properties can be set very simply and specifically for desired applications.

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The advantages of the invention can be seen in the simple operation and in the high activity and productivity of the carbonylation catalysts and the commercially available catalyst components.

The present invention also provides a process for preparing the catalyst used according to the present invention by mixing the components A and B. The invention further relates to the use of the catalyst in carbonylation reactions.

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Reagents

The reagents used came from Fluka, Aldrich or Merck and were used without further purification. The solvents were dried over molecular sieves and in each case degassed before use. The aluminum alkyl compounds were used as solutions in toluene.

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Analysis

NMR spectra were recorded on a Bruker AMX400 spectrometer. The IR measurements (KBr or directly as solution) were carried out on Bruker IFS 113V and IFS 66V instruments. For on-line IR studies to determine the reaction kinetics, a ReactIR™ (SiComp™ Dippersystem) from Mettler Toledo in a 250 ml Büchi reactor was employed.

General procedure:

The desired amount of epoxide (cf. Tables 1 to 4) was added to octacarbonyldicobalt $\text{Co}_2(\text{CO})_8$ (1 eq.) in diglyme at 0°C under an argon atmosphere. A compound of type B (1-6 eq.) was finally added (cf. Tables 1 to 4).

To charge the steel autoclave (100 or 250 ml), it was firstly evacuated and then charged under a countercurrent of argon. After the starting materials had been transferred to the steel autoclave, a carbon monoxide pressure of 10-65 bar was set and the carbonylation mixture was maintained at 75-105°C for a prescribed period. The carbonylation was stopped by reducing the pressure to ambient pressure, and the reaction solution obtained was cooled to 0°C, taken from the autoclave and analyzed. To separate off the catalyst, the resulting solution can be introduced into a mixture of diethyl ether/pentane. The catalyst and traces of polymer are separated off by filtration through silica gel, and subsequent fractional distillation of the filtrate gives the lactone in pure form.

Die Erfindung wird durch die nachstehenden Beispiele näher erläutert.

Examples

Example 1:

Octacarbonyldicobalt $\text{Co}_2(\text{CO})_8$ (260 mg) is dissolved in 16 ml of diglyme, the solution is cooled to 0°C and 8 ml of propylene oxide are added. After addition of 0.77 ml of 2N solution of Me_3Al in toluene, the reaction mixture is transferred in the absence of moisture and oxygen to a 100 ml steel autoclave provided with a glass liner. The carbonylation reaction is carried out at 75°C under 60 bar of CO for 5 hours. The carbonylation reaction is stopped by reducing the pressure to ambient pressure and cooling to 0°C. Analysis (^1H - and ^{13}C -NMR) of a sample indicates complete carbonylation of the epoxide and a lactone yield of > 95% (by-products are polyhydroxybutyrate and acetone).

Example 2:

In a 250 ml steel autoclave provided with an IR probe, octacarbonyldicobalt $\text{Co}_2(\text{CO})_8$ (780 mg) are dissolved in 50 ml of diglyme at 0°C under argon, and 26 ml of propylene oxide are added. After addition of 7 ml of a 2N solution of Me_3Al in toluene, the autoclave is pressurized with 60 bar of CO. The carbonylation reaction is carried out at 95°C under 60 bar of CO for 2 hours. The carbonylation reaction is stopped by reducing the pressure to ambient pressure and cooling to 0°C . Analysis (^1H - and ^{13}C -NMR) of a sample indicates complete carbonylation of the epoxide and a lactone yield of $> 95\%$ (by-products are polyhydroxybutyrate and acetone).

10 Example 3:

In a 250 ml steel autoclave provided with an IR probe, octacarbonyldicobalt $\text{Co}_2(\text{CO})_8$ (780 mg) are dissolved in 50 ml of diglyme at 0°C under argon, and 26 ml of propylene oxide are added. After addition of 7 ml of a 2N solution of Me_3Al in toluene, the autoclave is pressurized with 10 bar of CO. The carbonylation reaction is carried out at 75°C under 10 bar of CO for 4 hours. To stop the reaction, the pressure is brought down to ambient pressure and the mixture is cooled to 0°C . Analysis (^1H - and ^{13}C -NMR) of a sample indicates complete carbonylation of the epoxide and a lactone yield of $> 95\%$ (by-products are polyhydroxybutyrate and acetone).

20 Example 4:

Octacarbonyldicobalt $\text{Co}_2(\text{CO})_8$ (130 mg) is dissolved in 8 ml of diglyme, the solution is cooled to 0°C and 7 ml of butyloxirane are added. After addition of 0.39 ml of a 2N solution of Me_3Al in toluene, the reaction solution is transferred in the absence of moisture and oxygen to a 100 ml steel autoclave provided with a glass liner. The carbonylation reaction is carried out at 75°C under 60 bar of CO for 14 hours. The carbonylation reaction is stopped by reducing the pressure to ambient pressure and cooling to 0°C . Analysis (^1H - and ^{13}C -NMR) of a sample indicates about 70% carbonylation of the epoxide and a proportion of lactone in the product of $> 75\%$.

30 Example 5:

Octacarbonyldicobalt $\text{Co}_2(\text{CO})_8$ (130 mg) and tetraethylammonium tetracarbonylcobaltate $\text{Et}_4\text{NCo}(\text{CO})_4$ (232 mg) are dissolved in 10 ml of diglyme, the solution is cooled to 0°C and 6 ml of propylene oxide are added. After addition of aluminum isopropoxide $(i\text{-PrO})_3\text{Al}$, the reaction solution is transferred in the absence of moisture and oxygen to a 100 ml steel autoclave provided with a glass liner. The carbonylation reaction is carried out at 75°C under 60 bar of CO for 16 hours. The carbonylation reaction is stopped by reducing the pressure to ambient pressure and cooling to 0°C . Analysis (^1H - and ^{13}C -NMR)

of a sample indicates virtually complete carbonylation of the epoxide and a lactone yield of > 85% (by-products are polyhydroxybutyrate and acetone).

The following table summarizes further experiments which show that high conversions can be achieved in a short time using various combinations of cobalt compounds and component B.

Table 1: Variation of the Al component

| No. | Catalyst | Epoxide | Reaction conditions | Yield |
|-----|--|--------------|-----------------------------------|--------------------------------|
| 1 | Co ₂ CO ₈ (1 eq.) Me ₃ Al (2 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/ 5h | Conversion 100% Lactone 96% |
| 2 | Co ₂ CO ₈ (1 eq.) Et ₃ Al (2 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/5h | Conversion 100% Lactone 91% |
| 3 | Co ₂ CO ₈ (1 eq.) (i-Butyl) ₃ -Al (2 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/5h | Conversion 100% Lactone 87% |
| 4 | Co ₂ CO ₈ (1 eq.) Me ₃ Al (4 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/4h | Conversion 100% Lactone 92% |
| 5 | Co ₂ CO ₈ (1 eq.) Me ₃ Al (6 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/3h | Conversion 100% Lactone 88% |
| 6 | Co ₂ CO ₈ (1 eq.) EtCl ₂ Al (2 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/5h | Conversion 100% Lactone 85% |
| 7 | Co ₂ CO ₈ (1 eq.) Et ₂ ClAl ClEt ₂ Al (1 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/5h | Conversion 100% Lactone 90% |
| 8 | Co ₂ CO ₈ (1 eq.) MAO (2 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/20h | Conversion 90% Lactone 80% |
| 9 | Co ₂ CO ₈ (0.5 eq.) Et ₄ NCo(CO) ₄ (1 eq.) (i-PrO) ₃ Al (1 eq.) | PO (120 eq.) | 75°C/60 bar of CO/ diglyme/16h | Conversion 100% Lactone 87% |

All examples in Table 1 were carried out in a 100 ml steel autoclave provided with a glass liner; conversions and proportion of lactone were determined by NMR measurements on a sample; by-products are polyhydroxybutyrate and, in small amounts, acetone.

Table 2: Variation of the pressure

| No. | Catalyst | Epoxide | Reaction conditions | Yield |
|-----|---|--------------|----------------------------------|--------------------------------|
| 10 | Co ₂ CO ₈ (1 eq.) | PO (160 eq.) | 75°C/80 bar of CO/ diglyme/5h | Conversion 100% Lactone 90% |
| 11 | Co ₂ CO ₈ (1 eq.) | PO (160 eq.) | 75°C/60 bar of CO/ diglyme/5h | Conversion 100% Lactone 93% |
| 12 | Co ₂ CO ₈ (1 eq.) | PO (160 eq.) | 75°C/40 bar of CO/ diglyme/5h | Conversion 100% Lactone 92% |
| 13 | Co ₂ CO ₈ (1 eq.) | PO (160 eq.) | 75°C/20 bar of CO/ diglyme/5h | Conversion 100% Lactone 91% |
| 14 | Co ₂ CO ₈ (1 eq.) | PO (160 eq.) | 75°C/10 bar of CO/ diglyme/5h | Conversion 100% Lactone 92% |

All examples in Table 2 were carried out in a 250 ml steel autoclave without glass liner and with monitoring of the reaction by IR; conversions and proportions of lactone were determined by NMR measurements on a sample; by-products are polyhydroxybutyrate and, in small amounts, acetone.

Table 3: Variation of the temperature

| No. | Catalyst | Epoxide | Reaction conditions | Yield |
|-----|---|--------------|-----------------------------------|--------------------------------|
| 15 | Co ₂ CO ₈ (1 eq.) | PO (160 eq.) | 95°C/60 bar of CO/ diglyme/2h | Conversion 100% Lactone 92% |
| 16 | Co ₂ CO ₈ (1 eq.) | PO (160 eq.) | 105°C/60 bar of CO/ diglyme/1h | Conversion 100% Lactone 91% |

All examples in Table 3 were carried out in a 250 ml steel autoclave without glass liner and with monitoring of the reaction by IR.; conversions and proportions of lactone were determined by NMR measurements on a sample; by-products are polyhydroxybutyrate and, in small amounts, acetone.

Table 4: Variation of the epoxide/catalyst ratio

| No. | Catalyst | Epoxide | Reaction conditions | Yield |
|-----|---|---------------|-----------------------------------|--------------------------------|
| 17 | Co ₂ CO ₈ (1 eq.) | PO (300 eq.) | 75°C/60 bar of CO/ diglyme/10h | Conversion 100% Lactone 92% |
| 18 | Co ₂ CO ₈ (1 eq.) Me ₃ Al (4 eq.) | PO (600 eq.) | 75°C/60 bar of CO/ diglyme/7h | Conversion 100% Lactone 94% |
| 19 | Co ₂ CO ₈ (1 eq.) Me ₃ Al (4 eq.) | PO (1200 eq.) | 75°C/60 bar of CO/ diglyme/16h | Conversion 80% Lactone 90% |

All examples in Table 4 were carried out in a 100 ml steel autoclave provided with a glass liner; conversions and proportions of lactone were determined by NMR measurements on a sample; by-products are polyhydroxybutyrate and, in small amounts, acetone.

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